Effect of nonuniform hole-content distribution within the interlayer pair-tunneling mechanism of layered HTSC

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Abstract

The interlayer pair-tunneling (ILPT) mechanism for high-T_c superconductivity is able to predict the dependence of the (optimal) critical temperature T_c on the number of layers n within an homologous series of layered cuprate oxides. We generalize the mean-field procedure employed to evaluate T_c within an extended in-plane Hubbard model in presence of ILPT, developed for a bilayer complex ($n = 2$), to the case of $n = 3, 4$ *inequivalent* superconducting layers. As a function of doping, we show how a nonuniform hole-content distribution among different layers affects T_c. In particular, depending on doping, the onset of superconductivity may be ruled by inner or outer layers. The latter result may be related to recent experimental data of T_c as a function of pressure in Tl- and Bi-based layered superconductors.

Key words: High-T_c superconductivity; doping; layered cuprates; pressure effects.

1 Introduction

Among the features that have been recognized to characterize most high-T_c cuprate oxides are the dependences of the critical temperature T_c on hole-doping δ, and on the number n of CuO_2 layers in Bi-, Tl-, and Hg-based multilayered compounds. While $T_c = T_c(\delta)$ is usually well represented by a bell-shaped curve, T_c is seen to increase monotonically with increasing n, tending to a different saturation value for each homologous series for $n \gg 1$. While the former fact strongly supports the idea of a bidimensional pairing mechanism, the latter has suggested [1] that coherent tunneling of superconducting pairs between adjacent CuO_2 layers may considerably enhance T_c, and provide the in-plane order parameter $\Delta_{\mathbf{k}}$ with an unconventional anisotropy

in the Brillouin zone, its symmetry being determined solely by the nature and symmetry of the in-plane pairing [2]. A basic requirement is that the unconventional properties of the normal state suppress or greatly reduce coherent single-particle tunneling. This may be ensured by Anderson's orthogonality catastrophe or by spin-charge separation [1,2].

In order to single out the relevance of interlayer against purely bidimensional superconducting mechanisms, much theoretical as well as experimental effort has been devoted to the study of the two limiting cases of infinite-layered ($n = \infty$) compounds, and of single and bilayered compounds, with almost isolated layers. Another means to investigate the competition between 2D and interlayer effects is naturally provided by multilayered cuprates, with an intermediate number ($n = 2, 3$) of, but *inequivalent* layers.

Within the ILPT mechanism, one source of inequivalence among layers would be provided by the mechanism itself. In fact, a given CuO_2 layer may be coupled through ILPT to either one or two adjacent layers, depending on its position within a single unit cell. Therefore, one has to distinguish between inner and outer layers. This effect alone is able to account for the observed dependence of T_c on n [1]. Moreover, two layers are made inequivalent by their different position with respect to the 'reservoir' blocks at the edges of the unit cell, which induces a nonuniform distribution of hole-content among them. As said above, this is expected to affect bulk superconducting properties such as T_c.

In the following, we shall address the issue of the competition between these two sources of inequivalence in the case of a multilayered complex with a slightly nonuniform hole-content distribution.

2 The model

We consider the following model Hamiltonian for tightly bound interacting fermions in an n-layered complex [3]:

$$H = \sum_{\mathbf{k}\sigma\ell} \xi^{\ell}_{\mathbf{k}} c^{\ell\dagger}_{\mathbf{k}\sigma} c^{\ell}_{\mathbf{k}\sigma} + \sum_{\mathbf{k}\mathbf{k}'\ell\ell'} \tilde{V}^{\ell\ell'}_{\mathbf{k}\mathbf{k}'} c^{\ell\dagger}_{\mathbf{k}\uparrow} c^{\ell\dagger}_{-\mathbf{k}\downarrow} c^{\ell'}_{-\mathbf{k}'\uparrow} c^{\ell'}_{\mathbf{k}'\downarrow}. \tag{1}$$

Here, $c^{\ell\dagger}_{\mathbf{k}\sigma}$ ($c^{\ell}_{\mathbf{k}\sigma}$) creates (destroys) a fermion on layer ℓ ($\ell = 1, \ldots n$), with spin projection $\sigma = \uparrow, \downarrow$ along a specified direction, wave-vector $\mathbf{k}$ belonging to the first Brillouin zone (1BZ) of a two-dimensional (2D) square lattice, and band dispersion $\xi^{\ell}_{\mathbf{k}} = \varepsilon_{\mathbf{k}} - \mu^{\ell}$, measured relative to the chemical potential μ^{ℓ}. At variance with Ref. [3], here we explicitly assume that μ^{ℓ} may take different values depending on the layer index ℓ, thus accounting for different

hole content (band filling) n_h^ℓ on inequivalent layers [4]. Furthermore, on each layer we assume the 2D tight-binding dispersion relation (a being the lattice step):

$$\varepsilon_{\mathbf{k}} = -2t[\cos(k_x a) + \cos(k_y a)] + 4t' \cos(k_x a) \cos(k_y a), \tag{2}$$

where at least nearest-neighbours ($t = 0.25$ eV) as well as next-nearest neighbours ($t'/t = 0.45$) hopping has to be retained, in order to reproduce the most relevant properties common to the majority of the cuprate compounds [5].

In the Hamiltonian Eq. (1), interaction is restricted in the singlet channel only through:

$$\tilde{V}^{\ell\ell'}_{\mathbf{k}\mathbf{k}'} = \frac{1}{N} U_{\mathbf{k}\mathbf{k}'} \delta_{\ell\ell'} - T_J(\mathbf{k}) \delta_{\mathbf{k}\mathbf{k}'} (1 - \delta_{\ell\ell'}) \theta(1 - |\ell - \ell'|), \tag{3}$$

where $\theta(\tau)$ is the usual Heaviside step-function. The interaction term is thus made of an in-plane contribution $U_{\mathbf{k}\mathbf{k}'}$, which provides for Cooper pairing within each layer, and an *effective* contribution, arising from coherent pair-tunneling, here restricted between adjacent layers only. The ILPT matrix element $T_J(\mathbf{k})$ describes a second-order perturbation in the hopping matrix element $t_\perp(\mathbf{k})$ orthogonal to the CuO layers. Therefore, $T_J(\mathbf{k}) = t_\perp^2(\mathbf{k})/t$, where [2] $t_\perp(\mathbf{k})$ depends on $\mathbf{k}$ *locally* in the 1BZ as $t_\perp(\mathbf{k}) = t_\perp[\cos(k_x a) - \cos(k_y a)]^2/4$, as recently confirmed by detailed band-structure calculations [6], with $t_\perp = 0.1$—0.15 eV.

We shall not attempt at specifying the microscopic origin of the in-plane pairing mechanism. However, the potential $U_{\mathbf{k}\mathbf{k}'}$ has to be invariant under the symmetry transformations of the underlying crystal point group. Therefore, it may be expanded as a bilinear combination of basis functions for the irreducible representations of such group, viz. C_{4v} for the 2D square lattice. Such a series truncates after a finite number of terms, in the case of a finite-ranged potential. In Ref. [7] we have considered the competition among different symmetries arising from such a representation, in the presence of ILPT, as a function of band filling. Since we shall be mainly interested in the optimal filling region, where d-wave has been proved to prevail [7], in agreement with most experimental results, we may take $U_{\mathbf{k}\mathbf{k}'} = \lambda_3 g_3(\mathbf{k}) g_3(\mathbf{k}')$. Here, following the notation of Ref. [7], $g_3(\mathbf{k}) = \frac{1}{2}[\cos(k_x a) - \cos(k_y a)]$, and $\lambda_3 < 0$ is a phenomenological constant, related to in-plane inter-site coupling [7]. We remark that, in any case, no symmetry mixing would occur at exactly the critical temperature [7].

Standard mean-field (MF) treatment of Eq. (1) yields the BCS-like gap equation:

$$\Delta_{\mathbf{k}}^{\ell} = -\sum_{\mathbf{k}'\ell'} \tilde{V}_{\mathbf{k}\mathbf{k}'}^{\ell\ell'} \chi_{\mathbf{k}'}^{\ell'} \Delta_{\mathbf{k}'}^{\ell'}$$
$$= -\frac{1}{N} \sum_{\mathbf{k}'} U_{\mathbf{k}\mathbf{k}'} \chi_{\mathbf{k}'}^{\ell} \Delta_{\mathbf{k}'}^{\ell} + T_J(\mathbf{k}) \left[\chi_{\mathbf{k}}^{\ell+1} \Delta_{\mathbf{k}}^{\ell+1} + \chi_{\mathbf{k}}^{\ell-1} \Delta_{\mathbf{k}}^{\ell-1} \right], \tag{4}$$

where $\chi_{\mathbf{k}}^{\ell}$ denotes the pair susceptibility on layer ℓ. In the case of a multilayered complex, one usually introduces two more auxiliary gap functions $\Delta_{\mathbf{k}}^{\ell}$, identically vanishing, on the fictitious layers $\ell = 0$ and $\ell = n+1$ [3]. Eq. (4), supplemented by these conditions, explicitly displays two sources of inequivalence between layers. One source is the different value that μ^{ℓ} may assume on different layers. This is employed to describe a nonuniform hole-content distribution among layers. On the other hand, superconductivity in inner layers ($1 < \ell < n$) is enhanced by ILPT with two adjacent layers, whereas pairs in layers $\ell = 1$ and $\ell = n$, i.e. at the bottom and at the top of the n-layered stack, respectively, can tunnel coherently only towards one adjacent layer. It is known that both doping and the ILPT mechanism separately contribute to determine the critical temperature [7]. In the following, we shall study the interplay of these effects in the case of an n-layered complex.

In order to account for 'edge effects' [3], the Ansatz $\Delta_{\mathbf{k}}^{\ell} = \Delta_{\mathbf{k}} \sin(\ell\gamma)$ is introduced, thus allowing to decouple the $\mathbf{k}$-space from the ℓ-dependence of the order parameter [8]. The condition $\Delta_{\mathbf{k}}^{0} = \Delta_{\mathbf{k}}^{n+1} = 0$ then yields $\gamma = \frac{\pi}{n+1}$. Linearization of Eq. (4) at $T = T_c$ yields the following n nonlinear equations for the critical temperature T_c:

$$\frac{1}{\lambda_3} = -\frac{1}{N} \sum_{\mathbf{k}} g_3^2(\mathbf{k}) \frac{\chi_{\mathbf{k}}^{\ell}}{1 - T_J(\mathbf{k}) \hat{\chi}_{\mathbf{k}}^{\ell}}, \qquad \ell = 1, \dots n, \tag{5}$$

Here $\chi_{\mathbf{k}}^{\ell} = (2\xi_{\mathbf{k}}^{\ell})^{-1} \tanh(\frac{1}{2}\beta_c \xi_{\mathbf{k}}^{\ell})$ denotes the linearized pair susceptibility for layer ℓ. The renormalization factor, due to the ILPT mechanism [2], here contains the 'averaged' pair susceptibility

$$\hat{\chi}_{\mathbf{k}}^{\ell} = \left[\sin\left(\frac{\ell\pi}{n+1} \right) \right]^{-1} \left[\chi_{\mathbf{k}}^{\ell+1} \sin\left(\frac{(\ell+1)\pi}{n+1} \right) + \chi_{\mathbf{k}}^{\ell-1} \sin\left(\frac{(\ell-1)\pi}{n+1} \right) \right], \tag{6}$$

which reduces to the expression obtained in Ref. [3], when μ is the same for all layers.

For each $\ell = 1, \dots n$, Eq. (5) implicitly defines the critical temperature T_c^{ℓ} corresponding to the superconducting instability on layer ℓ. Eqs. (5) are decoupled with respect to ℓ, by virtue of the assumptions above, and because of linearization at T_c. However, the whole n-layered stack undergoes a metal-to-superconducting transition as soon as one of the n layers does, at least within this simplified MF approximation. Therefore, the largest of the solutions T_c^{ℓ}

of each of Eqs. (5) is to be regarded as the true critical temperature T_c.

One can straightforwardly recognize that the equation

$$\min_{\mathbf{k}}[1 - T_J(\mathbf{k})\hat{\chi}^{\ell}_{\mathbf{k}}] = 0, \qquad T_c = T^{\ell}_{\star}, \tag{7}$$

implicitly defines a lower bound to T^{ℓ}_c. In analogy with the bilayer case [7], Eq. (7) can be solved analytically for $T^{\ell}_{\star}$ as a function of μ^{ℓ}, showing that the ILPT mechanism actually *enhances* superconductivity at *all* band fillings. In the case of uniform hole-content distribution, $\mu^{\ell} = \mu$, all ℓ, near optimal doping Eq. (7) reduces to the known expression $T_{\star}/T_J = \frac{1}{2}\cos\frac{\pi}{n+1}$, where $T_J = t^2_{\perp}/t$, in qualitative agreement with the observed dependence of T_c on the number of layers n within the majority of layered HTCS [3].

3 Numerical results

Eqs. (5) have been solved numerically for T^{ℓ}_c as a function of μ^{ℓ} for $n = 3$ and $n = 4$. In these cases one may assume, for reasons of symmetry, that $\mu^1 = \mu^3 = \mu^{(o)}$, $\mu^2 = \mu^{(i)}$, and $\mu^1 = \mu^4 = \mu^{(o)}$, $\mu^2 = \mu^3 = \mu^{(i)}$, respectively, which define the hole-contents $n^{(i)}_h$ and $n^{(o)}_h$ within inner and outer layers. The following values of the parameters have been considered, in order to allow a qualitative comparison with the data for Bi- and Tl-based layered cuprates: $t_{\perp} = 0.08$ eV, $\lambda_3 = -0.2125$ eV [7].

Fig. 1 (left) shows T^{ℓ}_c as a function of $n^{(i)}_h$ and $n^{(o)}_h$ near optimal doping, in the case $n = 3$. In case $n = 4$, one obtains qualitatively similar results, not shown here. According to the above discussion, one can easily identify whether inner or outer layers give rise to superconductivity upon varying the hole-content distribution among layers. Along the contour $n^{(i)}_h = n^{(o)}_h$, corresponding to uniform hole-content distribution, T^{ℓ}_c is the same for all layers, and one recovers the typical bell-shaped dependence on doping. On the other hand, a shift from the uniform distribution makes either inner or outer layers prevail, depending on the actual values of $n^{(i)}_h$ and $n^{(o)}_h$.

Charge transfer from block 'reservoir' layers is generally agreed to provide the superconducting planes with a hole concentration δ, defined as the number of holes per unit CuO_2. This depends on the crystal structure, as well as on the chemical nature and formal valence of its constituents. Reservoirs may also act as 'buffers', i.e. they may repel charge excess, therefore confining holes within the CuO_2 layers. Moreover, oxygen relaxation processes may affect the hole-content distribution, due to the relatively high mobility of oxygen defects in the cuprate oxides, even at RT [9]. In multilayered cuprate compounds,

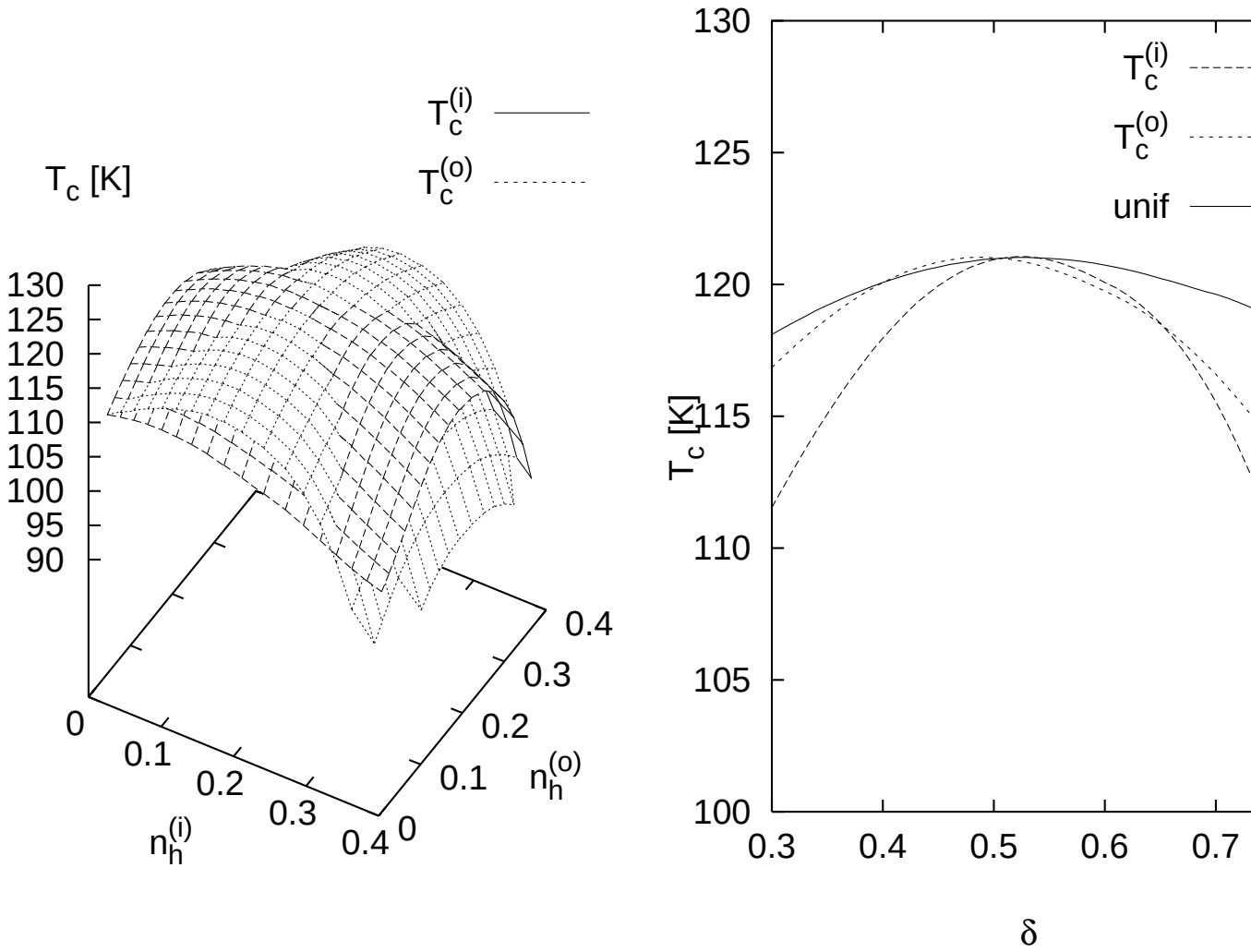

Fig. 1. *Left:* Critical temperatures $T_c^{(i,o)}$ for inner and outer layers in the case of an $n = 3$ layered complex, as functions of the hole-contents. *Right:* Same quantities, as functions of doping δ, for nonuniform hole-content distribution, as predicted by the point-charge model, with $\alpha_{\rm HT} = (m^\star/m_e)/\epsilon = 0.33$. The solid curve corresponds to the uniform distribution.

all these effects are believed to determine a nonuniform hole-content distribution among different layers, as evidenced experimentally [10]. Hydrostatic pressure is known to generally increase the overall hole-content as shown by Hall-resistance measurements in a variety of compounds [11], and may influence its distribution among layers as well.

Several models have been proposed in order to estimate the hole-contents $n_h^{(i)}$ and $n_h^{(o)}$ within inner and outer layers, respectively, as a function of doping δ [12–14]. Such models generally aim at minimizing the total carrier energy in the layered complex, expressed as a sum of band [13,12] or ionization [14] energy, plus electrostatic energy. In the *point-charge model* of Haines and Tal-

lon [13], the charge distribution within a given layer is realistically described as localized on the constituent ions of a lattice unit cell. For several compounds, the electrostatic energy can be therefore expressed as a Madelung sum [15]. Although the point-charge model is relatively system-dependent, its qualitative predictions depend mainly on the adimensional parameter $\alpha_{\rm HT} = (m^\star/m_e)/\epsilon$, expressing the ratio between the band-effective mass $m^\star$ in units of the electron bare mass m_e, with respect to the background dielectric constant ϵ.

According to the point-charge model, one finds that in $n = 3$ layered complexes the hole-content distribution is almost uniform, whereas for $n = 4$ the majority of carriers lie in the outer layers, yielding a vanishing $n_h^{(i)}$, especially at low doping δ [13,15]. As a result, more holes are available for the formation of pairs, and superconductivity is favoured in the outer layers. On the other hand superconducting pairs within inner layers may coherently tunnel towards *two* adjacent layers. Therefore, due to the ILPT mechanism, one expects a larger enhancement of T_c^ℓ within inner than within outer layers. As a function of doping δ, the two effects compromise, and a fairly complex scenario arise, as shown in Fig. 1 (right).

Both in the case $n = 3$ and $n = 4$ one observes that the onset of superconductivity is driven by outer layers at low doping, whereas inner layers rule out at higher doping, and particularly around optimal filling, yielding larger critical temperatures. Therefore, one may argue that the effect of the ILPT mechanism prevails near optimal doping. However, a second crossing may occasionally occurs at higher dopings, where superconductivity may be again dominated by outer layers.

The behaviour of $T_c = T_c(\delta)$ displayed in Fig. 1 (right) may eventually account for the doping contribution to the dependence of T_c on hydrostatic pressure P [16], assuming an approximately linear dependence of δ on P. The occurrence of 'crossovers' from $T_c^{(o)}$ to $T_c^{(i)}$ reminds one of the cusps experimentally exhibited by $T_c(P)$ in $n = 3$ Tl-2223 and $n = 4$ Tl-2234 [15]. Before a comparison could be made, however, intrinsic pressure effects should be realistically taken into account. In particular, the band structure parameters [16], as well as the ILPT amplitude and possibly the in-plane coupling constant, are expected to depend intrinsically on pressure.

4 Summary and concluding remarks

In conclusion, we have generalized a 2D extended Hubbard model in presence of ILPT mechanism for the case of a complex of n *inequivalent* layers. Different layers are made inequivalent by explicitly allowing a nonuniform hole-content distribution. The point-charge model has been employed to estimate the hole-

contents on inequivalent layers, for the simplest cases $n = 3, 4$. Moreover, the ILPT mechanism makes inner layers intrinsically different from outer layers, depending on the number (one or two) of adjacent layers. The critical temperature has been numerically evaluated at different band-fillings, in the mean-field approximation, for the simplified case of a decoupled dependence on $\mathbf{k}$ and ℓ of the order parameter. The competition between the enhancement due to the ILPT mechanism and the effect of a nonuniform hole-content distribution has been followed with respect to a variation of the overall hole-content δ. It is found that ILPT generally wins out around optimal doping, where inner layers are responsible of the superconducting transition. However, a crossover may occur at higher dopings, where outer layers may dominate. These results, although preliminary, are reminiscent of the nontrivial dependence of T_c on hydrostatic pressure observed in Tl-2223 and Tl-2234.

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